Relative Permittivity Measurements of Trifluoromethyl Methyl Ether and Pentafluoroethyl Methyl Ether

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The relative permittivities (ϵ_r) of trifluoromethyl methyl ether (HFE 143a) and pentafluoroethyl methyl ether (HFE 245mc) were measured as a function of temperature and pressure: (303 to 383) K and (1.5 to 31.6) MPa for HFE 143a and (313 to 323) K and (1.5 to 28.4) MPa for HFE 245mc. The relative permittivity was fitted to the reduced density (ρ_r) using the function ($\epsilon_r - 1$)/($2\epsilon_r + 1$). The apparent dipole moment (μ^*) of the liquid phase was calculated for each fluid using Kirkwood's theory of molecular polarizability.

Introduction

Chlorofluorocarbons (CFCs) were implicated as the primary agent responsible for ozone depletion. Current international agreements addressing global environmental issues (the Montreal and Kyoto Protocols) have provided the guidelines for effective phase-out of all ozone-depleting substances. Chlorofluorocarbons were used in a variety of applications including refrigeration, air conditioning, insulation manufacture, medical products, and cleaning of precision engineering components. It is therefore important for effective alternatives to be found.

Sekiya and Misaki¹ have investigated more than 150 fluorinated ethers as alternatives to CFCs for use as refrigerants, form-blowing agents, and cleaning solvents. Their results show that some hydrofluoroethers (HFEs) have low environmental impact, little or no flammability, low toxicity, and similar physical properties to CFCs. Further investigations for using HFEs as alternative refrigerants have been reported,^{2–4} and a short review regarding their use as CFC replacements was published by Bivens and Minor.⁵

While the environmental impact,^{6,7} critical properties,⁸⁻¹¹ thermal conductivities,^{12,13} vapor pressures,^{10,11,14} and other thermodynamic properties 15-17 of many fluoroethers have been studied, little work has been carried out to investigate the fundamental solvent properties of these fluids. Lagalante et al.¹⁸ measured the Kamlet-Taft thermosolvatochromic parameters of several HFEs and their azeotropic mixtures at (260 to 343) K. In addition, several Japanese research groups have measured the relative permittivities of a range of C₃ and higher fluorinated ethers^{19,20} (reported measurements do not include HFE 245mc). The relative permittivity (ϵ_r), often called the dielectric constant, is a macroscopic material property that is strongly related to molecular structure and can play an important role in solution properties.^{21,22} Measurement of ϵ_r has been shown to be a useful technique in characterizing molecular interactions and molecular ordering.^{23–25} In the current work we have measured ϵ_r of HFE 143a and HFE 245mc over a wide range of temperatures and pressures. The liquid-phase apparent dipole moments for these fluids are also reported for the first time.

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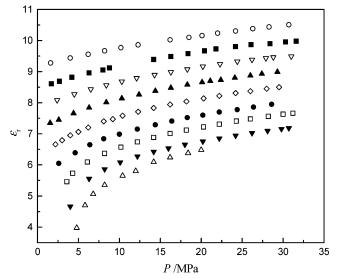


Figure 1. Pressure dependence of the relative permittivity (ϵ_r) of liquid and supercritical HFE 143a at various temperatures: \bigcirc , 302.8 K; \blacksquare , 313.2 K; \bigtriangledown , 322.8 K; \blacktriangle , 333.0 K; \diamondsuit , 343.4 K; \blacklozenge , 353.4 K; \square , 363.8 K; \blacktriangledown , 372.9 K; \triangle , 383.1 K.

Experimental Section

The relative permittivity was measured using a direct capacitance method. The cell capacitance in air (C_0) and the capacitance of the pure fluid (C) were measured, and ϵ_r was given by

$$\epsilon_{\rm r} = \frac{C}{C_{\rm o}} \tag{1}$$

Capacitances were measured with an applied potential of 1.0 V at 60.0 kHz using a Hewlett-Packard 4284A precision LCR meter (20 Hz to 1 MHz). The LCR meter was connected to an alternating six-plate capacitor using coaxial RG-316/U electrical leads (maximum working temperature 473 K) fitted with BNC connectors. The capacitor plate spacing was 1 mm, and each circular plate was 20 mm in diameter. The high-pressure electrical connectors were supplied by Sitec Sieber Engineering (Switzerland). The cell capacitance in air was measured to be 16.55 pF and was found to be independent of temperature. The pressure dependence of the cell constant was negligible over

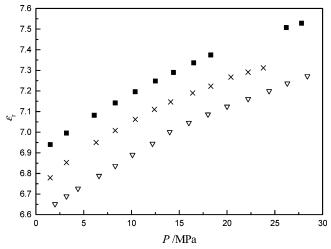


Figure 2. Pressure dependence of the relative permittivity (ϵ_i) of liquid HFE 245mc at various temperatures: **I**, 312.9 K; ×, 318.0 K; \bigtriangledown , 323.4 K.

the pressure ranges studied ($< 7 \times 10^{-5}$ % variation in $C_{\rm o}$). Temperature was monitored using a Type K ServoTech thermocouple (\pm 0.5 K). Pressure measurements have an associated uncertainty of \pm 0.2 MPa. The experimental procedure and equipment validation has been described in detail elsewhere.²⁶

Each data point is the average of at least three replicate measurements, with maximum standard deviation (σ) for a given condition over all temperatures and pressures studied = 0.011.

Each replicate measurement was made by refilling the capacitance cell with the fluid under investigation. The relative permittivity values have an expanded uncertainty of \pm 7.4 % (95 % confidence level).

The HFE 143a and HFE 245mc gases were supplied by ABCR GmbH (Germany) with a stated purity of (99.0 and 98.0) % by volume, respectively. Each was used as received without further purification. Impurities as stated by certificate of analysis were HFE 143a: dimethyl ether (CH₃OCH₃) and chloromethane (CH₃Cl); HFE 245mc: acetyl fluoride (CF₃COF) and methyl trifluoroacetate (CF₃COOCH₃).

Results

Figures 1 and 2 show the relative permittivity of HFE 143a and HFE 245mc as a function of temperature and pressure compiled from the data in Tables 1 and 2. The relative permittivity increases with increasing pressure and decreasing temperature. Hence, the isothermal density dependence, $(\delta \epsilon_r / \delta \rho)_T$, of ϵ_r is always positive and the isobaric temperature dependence, $(\delta \epsilon_r / \delta T)_P$, is always negative.

The observed temperature-dependent behavior is due to the presence of a dipole in the HFE molecules. As temperature is increased, the molecular thermal motion increases and the ability of the molecules to orient and align their dipoles with the applied electric field is reduced. This reduced alignment of dipoles leads to a reduction in ϵ_r at higher temperatures.

Table 1. Relative Permittivity (ϵ_r) Values for Liquid and Supercritical HFE 143a at (303 to 383) K^a

P/MPa	$\epsilon_{\rm r}$	$ ho_{\rm cal}/{\rm kg}{ m \cdot}{\rm m}^{-3}$	P/MPa	$\epsilon_{\rm r}$	$ ho_{\rm cal}/{\rm kg}{ m \cdot}{\rm m}^{-3}$	P/MPa	$\epsilon_{\rm r}$	$ ho_{\rm cal}/{\rm kg}{ m \cdot}{\rm m}^{-3}$	P/MPa	$\epsilon_{\rm r}$	$ ho_{\rm cal}/{\rm kg}{ m \cdot}{\rm m}^{-3}$	P/MPa	$\epsilon_{\rm r}$	$ ho_{ m cal}/ m kg\cdot m^{-3}$
	T/K = 302.8		T/K = 313.2		T/K = 322.8		T/K = 333.0		T/K = 343.4					
1.6	9.28	1086.0	1.7	8.61	1046.2	2.4	8.08	1015.7	1.5	7.35	965.8	2.2	6.66	914.5
4.0	9.44	1098.3	2.7	8.69	1052.8	4.5	8.27	1031.9	2.6	7.45	975.4	3.0	6.79	928.5
6.2	9.56	1107.3	4.2	8.82	1063.2	6.5	8.43	1045.1	4.4	7.66	994.8	4.0	6.95	945.0
8.2	9.67	1115.4	6.3	8.96	1074.2	8.4	8.56	1055.4	6.3	7.84	1010.7	5.0	7.06	956.0
10.2	9.77	1122.6	8.0	9.05	1081.1	10.3	8.68	1064.7	8.4	8.01	1025.0	6.2	7.20	969.5
12.3	9.86	1128.9	8.8	9.12	1086.4	12.1	8.79	1073.0	10.3	8.14	1035.6	8.1	7.40	987.9
16.2	10.02	1140.0	14.2	9.39	1106.0	14.3	8.89	1080.4	12.4	8.26	1045.2	9.2	7.47	994.2
18.5	10.10	1145.3	16.3	9.49	1113.0	16.3	8.98	1086.9	14.4	8.38	1054.4	10.4	7.58	1003.8
20.1	10.16	1149.3	18.4	9.58	1119.1	18.3	9.07	1093.3	16.3	8.48	1061.9	12.3	7.71	1014.7
22.0	10.24	1154.6	20.4	9.67	1125.2	20.2	9.16	1099.5	18.3	8.58	1069.3	14.2	7.83	1024.6
24.2	10.31	1159.1	21.8	9.73	1129.2	22.3	9.24	1105.0	20.1	8.66	1075.1	16.2	7.94	1033.4
26.3	10.38	1163.6	24.2	9.81	1134.4	24.4	9.32	1110.4	21.1	8.70	1077.9	18.2	8.04	1041.2
28.3	10.44	1167.3	26.1	9.87	1138.3	26.2	9.38	1114.4	22.4	8.73	1080.0	20.3	8.14	1048.8
30.7	10.51	1171.7	28.4	9.90	1140.2	27.9	9.43	1117.6	24.1	8.80	1084.9	22.2	8.22	1054.7
			30.3	9.95	1143.4	28.8	9.46	1119.6	26.0	8.87	1089.7	24.2	8.31	1061.3
			31.6	9.98	1145.3	31.0	9.49	1121.5	27.4	8.93	1093.8	26.0	8.37	1065.6
									29.3	8.99	1097.8	28.0	8.45	1071.2
												29.5	8.50	1074.7
	T/K = 353.4		T/K = 363.8		T/K = 372.9		$T/K = 383.1^{b}$							
2.6	6.05	864.8	3.6	5.46		4.0	4.66		4.8	3.97				
4.6	6.39	904.6	4.3	5.73		6.3	5.55		5.8	4.70				
6.4	6.65	932.5	6.1	6.09		8.2	5.86		6.8	5.06				
8.1	6.84	951.6	8.2	6.37		10.1	6.08		8.2	5.35				
10.0	6.99	966.0	10.2	6.57		12.2	6.27		10.4	5.65				
12.2	7.15	980.7	12.2	6.74		14.2	6.42		12.0	5.80				
14.4	7.29	993.2	14.2	6.88		16.0	6.53		14.2	6.09				
16.4	7.41	1003.4	16.2	7.01		18.0	6.65		16.2	6.24				
18.3	7.52	1012.6	18.2	7.12		20.2	6.75		18.4	6.38				
20.2	7.60	1019.1	20.3	7.22		22.2	6.85		20.0	6.48				
22.2	7.70	1027.1	22.2	7.31		24.2	6.94							
24.4	7.78	1033.3	24.3	7.41		26.2	7.02							
26.4	7.86	1039.4	26.0	7.48		28.1	7.09							
28.6	7.95	1046.1	28.2	7.56		29.8	7.15							
			30.0	7.63		30.7	7.18							
			31.2	7.66										

^a Liquid densities (ρ_{cal}) at (303 to 353) K were calculated using eq 2. ^b Supercritical.

Table 2. Relative Permittivity $(\epsilon_{\rm r})$ Values for Liquid HFE 245mc at (313 to 323) ${\rm K}^a$

Р		$ ho_{ m cal}$	Р		$ ho_{ m cal}$	Р		$ ho_{ m cal}$	
MPa	$\epsilon_{\rm r}$	kg•m ⁻³	MPa	$\epsilon_{\rm r}$	kg•m ⁻³	MPa	$\epsilon_{\rm r}$	kg•m ⁻³	
T	K = 3	12.9	T/K = 318.0			T/K = 323.4			
1.5	6.94	1203.2	1.5	6.78	1186.2	2	6.65	1174.4	
3.2	7.00	1213.2	3.2	6.85	1198.2	3.2	6.69	1181.4	
6.1	7.08	1226.3	6.3	6.95	1215.0	4.4	6.73	1188.4	
8.3	7.14	1236.0	8.3	7.01	1224.8	6.6	6.79	1198.7	
10.4	7.20	1245.5	10.4	7.06	1232.9	8.3	6.84	1207.2	
12.5	7.25	1253.3	12.4	7.11	1240.9	10.1	6.89	1215.5	
14.4	7.29	1259.4	14.1	7.15	1247.2	12.2	6.94	1223.8	
16.5	7.34	1267.1	16.4	7.19	1253.5	14	7.00	1233.5	
18.3	7.37	1271.6	18.3	7.22	1258.1	16	7.05	1241.5	
26.2	7.51	1292.3	20.4	7.27	1265.8	18	7.09	1247.8	
27.8	7.53	1295.2	22.2	7.29	1268.8	20	7.12	1252.5	
			23.8	7.31	1271.9	22.2	7.16	1258.7	
						24.4	7.20	1264.9	
						26.3	7.24	1271.0	
						28.4	7.27	1275.5	

^{*a*} Liquid densities (ρ_{cal}) at (313 to 323) K were calculated using eq 2.

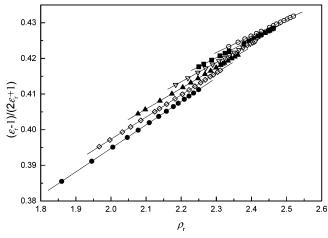


Figure 3. Linear dependency of $(\epsilon_r - 1)/(2\epsilon_r + 1)$ on the reduced density (ρ_r) of liquid HFE 143a. The standard deviation (σ) for each linear correlation is given in parentheses: \bigcirc , 302.8 K (3.3 × 10⁻⁶); \blacksquare , 313.2 K (3.6 × 10⁻⁶); \bigtriangledown , 322.8 K (2.6 × 10⁻⁶); \blacktriangle , 333.0 K (2.6 × 10⁻⁶); \diamondsuit , 343.4 K (2.8 × 10⁻⁶); \blacklozenge , 353.4 K (3.2 × 10⁻⁶).

As carried out previously for hydrofluorocarbon solvents,²⁷ the relative permittivity data were fitted to the empirical equation:

$$\frac{\epsilon_{\rm r} - 1}{2\epsilon_{\rm r} + 1} = A + B\rho_{\rm r} \tag{2}$$

where ρ_r is the reduced density ($\rho_r = \rho/\rho_c$, where ρ is the density at given temperature and pressure conditions and ρ_c is the critical density) and A and B are temperature-dependent constants. Fitting was carried out using literature¹⁵ liquid densities from (303 to 353) K for HFE 143a and from (313 to 323) K for HFE 245mc. The A and B constants for HFE 143a and HFE 245mc are reported in Table 3 along with their critical parameters.^{8,10} The correlations have a maximum deviation in density, [(calculated – literature)/calculated] \times 100, of \pm 0.42 % for HFE 143a and of \pm 0.26 % for HFE 245mc. The function ($\epsilon_{\rm r}$ – 1)/($2\epsilon_r + 1$) varies linearly with the isothermal reduced density (Figures 3 and 4), and the constants of proportionality vary linearly with the reduced temperature $(T_r = T/T_c, \text{ where } T \text{ is})$ the temperature at which ϵ_r is measured and T_c is the critical temperature) (Figures 5 and 6). Liquid densities (ρ_{cal}) for HFE 143a from (303 to 353) K and (1.5 to 31.6) MPa and HFE

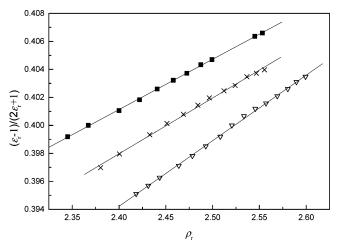


Figure 4. Linear dependency of $(\epsilon_r - 1)/(2\epsilon_r + 1)$ on the reduced density (ρ_r) of liquid HFE 245mc. The standard deviation (σ) for each linear correlation is given in parentheses: \blacksquare , 313.2 K (5.1 × 10⁻⁵); ×, 318.0 K (1.3 × 10⁻⁴); \bigtriangledown , 323.4 K (9.4 × 10⁻⁵).

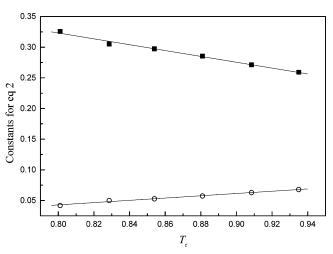


Figure 5. Linear dependency on reduced temperature (T_t) for the constants of proportionality of eq 2 for HFE 143a. The standard deviation (σ) for each linear correlation is given in parentheses: \blacksquare , A (0.003); \bigcirc , B (0.001).

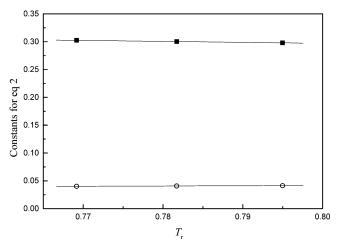


Figure 6. Linear dependency on reduced temperature (T_r) for the constants of proportionality of eq 2 for HFE 245mc. The standard deviation (σ) for each linear correlation is given in parentheses: \blacksquare , A (1.2×10^{-5}); \bigcirc , B (2.8×10^{-5}).

245mc from (313 to 323) K and (1.5 to 28.4) MPa calculated using eq 2 are reported in Tables 1 and 2, respectively.

Table 3. Critical Properties of HFE 143a¹⁰ and HFE 245mc⁸ and the A and B Constants for Equation 2, Where T_r is the Reduced Temperature

	$T_{\rm c}/{ m K}$	P _c /MPa	$ ho_{ m c}/{ m kg}\cdot{ m m}^{-3}$	Α	В
HFE 143a HFE 245mc	378.0 406.8	3.68 2.89	439 499	$\begin{array}{l} 0.626631 - 0.389281 T_{\rm r} \\ 0.434909 - 0.17232 T_{\rm r} \end{array}$	$\begin{array}{c} 0.138009T_{\rm r}-0.066685\\ 0.006248+0.044087T_{\rm r} \end{array}$

At 294.15 K and atmospheric pressure the National Institute of Advanced Industrial Science and Technology (AIST) report the relative permittivities of HFE 143a and HFE 245mc to be 9.18 and 7.26, respectively.²⁸ The liquid densities of HFE 143a and HFE 245mc at 294.15 K and atmospheric pressure were calculated according to Kayukawa et al.¹⁵ Equation 2 was used to calculate ϵ_r under these conditions, yielding values of 9.64 and 7.65 for HFE 143a and HFE 245mc, respectively. These values are slightly greater than those reported by AIST with a deviation of + 5.0 % for HFE 143a and + 5.4 % for HFE 245mc. Although the deviation is within the \pm 7.4 % expanded uncertainty reported in the current work, no experimental details for measurement of the AIST values are given, and the reason for the deviation is not apparent.

The apparent dipole moment (μ^*) of a polar liquid can be related to ϵ_r through the equation²⁷

$$KF = \frac{(\epsilon_r - 1)(2\epsilon_r + 1)}{9\epsilon_r} \left(\frac{3M}{N_A\rho}\right) = \left(\alpha + \frac{\mu^{*2}}{3\epsilon_o k_BT}\right)$$
(3)

where *M* is the molar mass, N_A is the Avogadro constant, α is the molecular polarizability, ϵ_0 is the vacuum permittivity, and k_B is the Boltzmann constant.

By performing a linear regression as a function of 1/T, the value of μ^* can be determined. The Kirkwood function (KF) was approximately independent of density at each temperature. The mean KF is plotted as a function of 1/T in Figure 7, yielding μ^* values of $\{(1.20 \pm 0.15) \times 10^{-29}\}$ C·m for HFE 143a and of $\{(8.41 \pm 1.08) \times 10^{-30}\}$ C·m for HFE 245mc. The expanded uncertainties in the dipole moment values are based on the expanded uncertainties associated with the density values (\pm 0.024 %),¹⁵ temperature measurements (\pm 0.6 K), and relative permittivity values (\pm 7.4 %). The confidence level of the expanded uncertainties is 95 %.

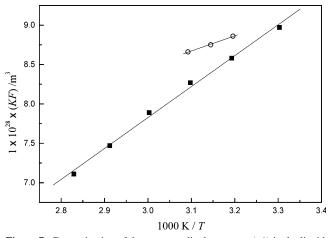


Figure 7. Determination of the apparent dipole moment (μ^*) in the liquid phase by the Kirkwood representation. The standard deviation (σ) for each linear correlation is given in parentheses: \blacksquare , HFE 143a (5.2×10^{-30}); \bigcirc , HFE 245mc (9.1×10^{-31}).

Conclusions

The relative permittivities of HFE 143a from (303 to 383) K and (1.5 to 31.6) MPa and of HFE 245mc from (313 to 323) K and (1.5 to 28.4) MPa have been measured. Under these conditions the isothermal pressure dependence of the relative permittivity, $(\delta \epsilon_r / \delta P)_T$, is always positive, and the isobaric temperature dependence, $(\delta \epsilon_r / \delta T)_P$, is always negative. The relative permittivity values have been fitted to the reduced density using the function $(\epsilon_r - 1)/(2\epsilon_r + 1)$. At pressures above the saturation curve this function can be used to estimate the liquid densities of HFE 143a over the temperature range (303 to 353) K and HFE 245mc over the temperature range (313 to 323) K to within ± 0.42 % and ± 0.26 % of experimental values, respectively. The apparent dipole moment of both fluids in the liquid phase is also reported for the first time.

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